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INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

(51) International Patent Classification 3 : G01N 27/46

A1

(11) International Publication Number: WO 83/ 01687

(43) International Publication Date: 11 May 1983 (11.05.83)

(21) International Application Number: PCT/AU82/00175

(22) International Filing Date: 27 October 1982 (27.10.82)

(31) Priority Application Number: PF 1334

(32) Priority Date: 29 October 1981 (29.10.81)

(33) Priority Country: AU

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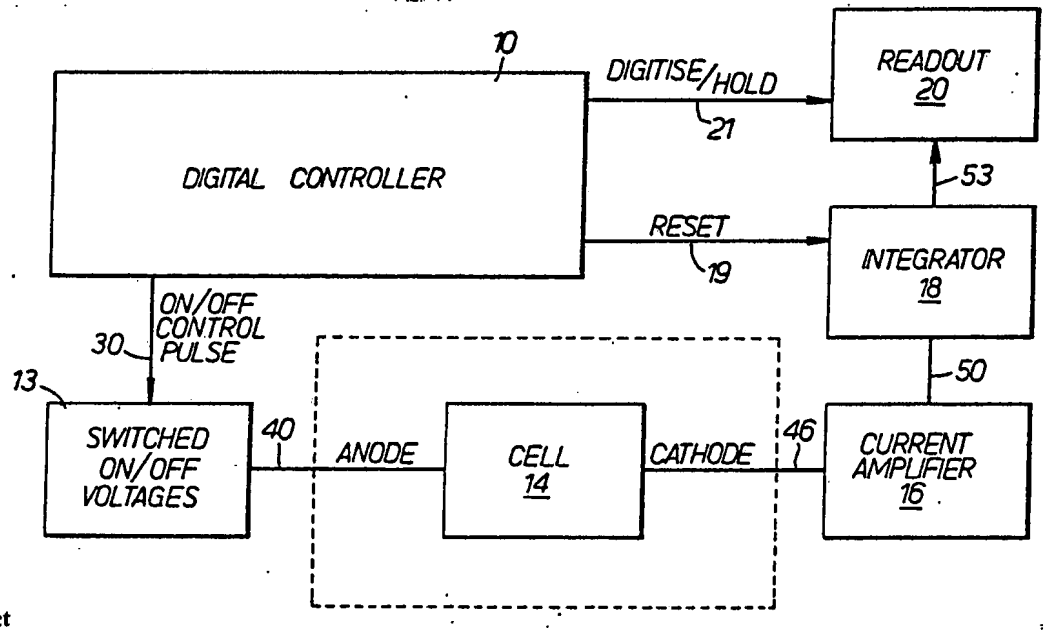
(81) Designated States: AT (European patent), AU, BE (European patent), CH (European patent), DE (European patent), FR (European patent), GB (European patent), JP, LU (European patent), NL (European patent), SE (European patent), US.

Published
With international search report.

(54) Title: PULSED ELECTRO-ANALYSIS



(57) Abstract

Measurement of the concentration of an electroactive material (analyte) in an electrolyte is effected by forming a cell (14) of the electrolyte and applying pulses of an On-potential to the anode of the cell (14). The current through the cell (14) is measured and integrated for the time that the On-potential is applied and at least part of the time period between the application of the pulses On-potential. A controller (10) controls the lenght of the on-period and the pulses repetition frequency. The integrated cell current is preferably displayed in digital form on a readout device (20), which may also provide a record of its output. A typical application of the invention is the measurement of the concentration of oxygen in water.

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TITLE: "PULSED ELECTRO-ANALYSIS"

FIELD OF THE INVENTION

This invention concerns electro-analysis. That is, it concerns measurement of the concentration of an electroactive material (termed an "analyte") in an electrolyte. The invention was developed primarily to measure the concentration of oxygen dissolved in water but, as will be seen, the invention is not limited to this application.

BACKGROUND ART

The apparatus currently used to measure the analyte concentration in an electrolyte consists of an electrolytic cell with electrical connections to an instrument which contains cell potential control circuitry and electrode current measuring means. That instrument provides an indication of the analyte concentration. Examples of electrolytic cells that are suitable for these measurements are voltammetric and galvanic oxygen sensors, and dropping mercury electrode cells. Generally, those cells contain an indicator electrode and a counter electrode, and sometimes a third electrode which is used as a separate reference electrode. All the electrodes are in contact with the electrolyte under investigation. Electrode current, which is related to analyte level in the vicinity of the indicator electrode, is that current which flows between the indicator electrode and the external circuit. Cell potential (or indicator electrode potential) is the potential of the indicator electrode relative to whatever electrode is being used as a reference electrode.

As indicated above, the present invention, although applicable to all types of electrolytic cells and sensors, has a particular application to the measurement of dissolved oxygen level (partial pressure) in still water. That measurement involves the

use of membrane-covered oxygen sensors. Because that application of the present invention will be referred to frequently in this specification, it is convenient to use the terms "cathode" and "anode" for "indicator electrode" and "counter electrode", respectively, when the measurement of dissolved oxygen is discussed.

In the measurement of steady-state oxygen level (with a constant cell potential imposed by the voltage source of the measuring instrument) using these sensors, errors in the measurements arise from (a) depletion of the oxygen level in the vicinity of the cathode if the liquid oxygen-containing sample is not flowing, and (b) a high non-linear temperature coefficient. It is known that these errors may be nearly offset by applying an oxygen-reducing cell potential in pulses.

The most common prior art pulse method that is used with oxygen sensors, whether or not the cathode is membrane-covered, is pulse amperometry (sometimes called "chronoamperometry" or "pulse polarography"), and with this technique it has been recognised that unacceptably high "zero" readings at zero oxygen content are generally observed. This shortcoming appears to be related to a substantial capacitive current flowing at the end of the pulse due to a change in the double layer capacity of the cathode when its potential is changed. Faradaic current (that is, current related to oxygen concentration) cannot be measured directly as it occurs with a component of capacitive current which has a magnitude which depends on the cell design and the time elapsed since the start of the pulse.

Another pulse technique has been reported to produce low "zero" errors, but each measurement in that technique requires a large number of pulses, and the calibration of the instrument was reported to be non-linear.

DISCLOSURE OF THE PRESENT INVENTION

It is an objective of the present invention to provide pulsed electro-analysis apparatus which includes means to cancel the effect of capacitive current, and thus enables accurate measurement to be made of the Faradaic current flowing during each pulse.

This objective is achieved by integrating the cell current for a period which encompasses the time during which the applied pulse is "on" and at least part of the "off" time before the next pulse.

According to the present invention, there is provided an instrument for measuring the concentration of electroactive material, said instrument being adapted for use with an electrolytic cell and comprising means to apply sequentially to the cell an On-potential for an On-period of time and an Off-potential for an Off-period of time, characterised in that said apparatus includes

a) integration means to integrate the cell current during the application of the On-potential and the Off-potential for a period of time comprising the On-period and at least part of the Off-period;

b) control means to control the length of the On-period and the Off-period and to preselect the values of the On-potential and the Off-potential; and

c) measuring means to display and/or record the integrated cell current.

The present invention also encompasses a method of electro-analysis to determine the concentration of electroactive material in an electrolytic cell, said method comprising sequentially applying an On-potential to the cell for an On-period of time and applying an Off-potential for an Off-period of time, characterised by the steps of

a) selecting said On-potential to produce a measurable Faradaic cell current and selecting said Off-potential to produce zero Faradaic current in said cell; and

b) integrating the total cell current during the application of the On-potential and the Off-potential for a period of time comprising the On-period and at least part of the Off-period, so that the capacitive component of the integral of the On-period cell current is cancelled by the integration of the Off-period cell current.

The criteria used to select the values of the On- and Off-potentials, and the On- and Off-periods are the same criteria that are used in the existing pulse techniques (for example, pulse amperometry, pulse polarography or pulse coulometry), with the exception that there is no need for capacitive current to be very low at the end of a pulse (that is, at the end of an On-period) in order to obtain very low "zero" readings. Thus, other factors being equal, use of the present invention permits lower "zero" readings and narrower pulses.

In accordance with the method of the present invention, the On-potential is selected to produce a measurable Faradaic current while the Off-potential is selected to produce zero Faradaic current. When these selected potentials are applied to the cell, the electrode current is integrated over an integration

period which comprises the total On-period and at least that part of the Off-period which is sufficient for cancellation of the capacitive component of the integral of the On-period electrode current. This is achieved if the integral of Off-period current is equal in magnitude but opposite in sign to the integral of On-period capacitive current. The minimum value of this integration period, which should be determined experimentally, depends on the type of electrolytic cell used in conjunction with the instrument, and also on the pulse regime employed by the instrument.

The present invention may be used in applications where the Faradaic Off-potential current is not zero and its integral is not small in relation to the On-period Faradaic current integral. Integration under these conditions would normally produce an error in the indication of the analyte level. The instrument and method of the present invention may be used to cancel this error by instrument circuitry which provides a first integral for a first time period corresponding to an integration period as stated in the last preceding paragraph, and then subtracts from it a second integral produced by integrating the Off-period current for a second time period beginning after the capacitive component has decayed to an insignificant value. These two time periods are equal in length. Examples of instances where this approach will be adopted are the measurement of low analyte concentration and the cancellation of the effects of offset voltages or currents produced by the instrument electronic circuitry.

Having portrayed the general nature of the present invention, preferred embodiments thereof will now be described by way of illustration. In the following description, reference will be made to the accompanying drawings.

BRIEF DESCRIPTION OF THE DRAWINGS

Figure 1 depicts a typical oxygen sensor voltammogram.

Figure 2 is a graph of cathode current versus time for pulsed operation of an electrolytic cell.

Figure 3 is a timing diagram for the operation of the instrument of Figure 4.

Figure 4 is a block diagram of the preferred embodiment of the present invention for use in the analysis of the oxygen content of still water.

Figure 5 is a circuit diagram of one form of On/Off voltage source and switching circuit.

Figure 6 is a circuit diagram of an alternative means of providing the On/Off voltage.

Figure 7 is a circuit diagram of a digital integrator.

DETAILED DESCRIPTION OF THE ILLUSTRATED EMBODIMENTS

Figure 1 shows a voltammogram obtained by plotting equilibrium cathode current against the cathode potential (relative to anode) for a constant oxygen level. The voltammogram may conveniently be used to select the On- and Off-potentials for the present invention. Typically, the potentials shown in Figure 1 as A and B, respectively, would be selected.

Figure 2 is a graphical representation of the cathode current that is produced by an electrolytic cell of which the cathode potential is controlled by the On/Off voltage source of the instrument of the invention. In Figure 2, line D represents the total electrode current including the Faradaic component and the capacitive component shown by line C. It will be noted that the Off-period Faradaic current is practically zero (that is, it is very small compared with the On-period Faradaic current). Cancellation of

the capacitive component may be achieved if the integration period begins at time T1 and ends at (or subsequent to) time T3.

The instrument depicted by the block diagram of Figure 4 includes a digital controller 10, one output of which is connected by line 30 to control a switched On/Off voltage source 13. Source 13 sequentially applies preselected On and Off voltages to the anode of cell 14 via line 40. The cathode current of cell 14 is connected to a current amplifier 16 via line 46. Amplifier 16 has a voltage output which is proportional to the cathode current. This voltage is integrated by the integrator 18 and the integrated value is displayed by the readout device 20.

The digital controller 10 resets the integrator 18 via line 19 immediately prior to the beginning of each On-period and also provides signals via line 21 to the readout device at the end of the predetermined integration period to cause it to digitise (if necessary) and display the integrated current measurement. The durations of the On- and Off-periods and the integration period are controlled by the digital controller according to a preselected program. The On- and Off-potentials, applied by the switched On/Off voltage source 13, are also preselected by the operator of the instrument.

One form of circuit that may be used for the switched On/Off voltage source 13 is shown in Figure 5. In this circuit, there are an On-voltage source 38 and an Off-voltage source 52, with two electronic switch elements 32 and 34 arranged in series with the voltage sources 38 and 52 respectively. The switch elements apply selected voltages via series resistor R1, through a buffer amplifier circuit 44, to line 40, which is connected to the anode of the cell 14. The control line 30 from the digital controller 10 is connected directly

to switch 32 and is also connected, via inverter 36, to switch 34. Switch 32 is normally off whilst switch 34 is normally on, so that, in the absence of a signal from the controller, the Off-voltage is fed to line 40 via the buffer amplifier 44. Upon the receipt (through line 30) of a pulse from the controller 10 (the duration of which pulse is the On-period), the action of inverter 36 ensures that switch 34 is turned off and switch 32 is turned on to connect the On-voltage source 38 via the buffer amplifier 44 to line 40.

A second example of the switched voltage source 13 is shown in Figure 6. This form of the source comprises a voltage reference 31, an amplifier 45 with series resistor 37 permanently connected and a resistor 35 connected during the On-period by switch 33. The output of amplifier 45 is connected to line 40, which (as already shown) is connected to the anode of the cell 14. Control line 30 from the digital controller 10 is connected to switch 33, to which the reference voltage 31 is also connected. Switch 33 is normally off so that in the absence of a signal on line 30 from the controller, the amplifier 45 feeds the Off-voltage to line 40, the Off-voltage being set by series resistor 37 and feedback resistor 39. Upon receipt of a pulse (through line 30) from the controller, the duration of which is the On-period, switch 33 is turned on, thereby connecting resistor 35 in parallel with series resistor 37. In this situation, amplifier 45 applies the On-voltage to line 40, the On-voltage being set by resistors 35 and 37 in parallel, together with feedback resistor 39. If a negative Off-voltage should be required (for example, for a galvanic oxygen sensor), resistor 37 will be connected to a different (positive) voltage source.

Whether the switched voltage source of Figure 5 or the source of Figure 6 is used, the On- and Off-voltages are applied sequentially to line 40, which is connected to the anode of cell 14. The cathode of cell 14 is connected via line 46 to a virtual ground (zero volt) input of the current amplifier 16. Thus the On- and Off-voltages applied by the instrument (see Figures 3 to 7) are potentials of the anode relative to the cathode, these voltages being opposite in sign to the cathode potential shown in Figure 1.

The integrator 18 may be a simple analog integrator.

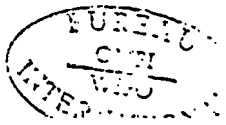
The circuit shown in Figure 7 includes a digital integrator as an example of integrator 18. Briefly, the circuit of Figure 7 provides an up digital count for positive integrands and subtracts down counts from the up count for negative integrands. In more detail, this circuit comprises a switchable unity gain circuit 58 which switches positive voltages on line 50 directly to line 51 and inverts negative voltages from line 50 to appear as positive voltages on line 51. These voltages are presented to the input of a unipolar voltage-to-frequency converter 66 which provides, at line 67, a pulse output having a frequency which is proportional to the magnitude of the input voltage. The up/down counter 68 converts the frequency input at line 67 into a digital count proportional to frequency. The counter 68 is controlled by a signal from the digital controller introduced by line 30, which causes the counter to count up for positive voltages at line 50 and to count down for negative voltages at line 50. The counter is reset by the digital controller via line 19 prior to the beginning of the next On-period. At the end of each integration period, the integrated count

for this period, as determined by counter 68, is displayed in readout device 20, controlled by a signal on line 21.

The timing diagram shown in Figure 3 provides an illustration of the working of the instrument of the present invention. It contains various graphs of instrument and indicator electrode currents and voltages plotted against a common time events scale, to show the events controlled by the digital controller 10.

Graphs 40a and 46a of Figure 3 depict, respectively, the voltage applied to the anode and the corresponding cathode current. The reset pulse of control signals 19 resets the integrator so that the integration period commences at the beginning of the On-period. Graph 53 shows the output of integrator 18. The end of the integration period is controlled from the digital controller by pulse on line 21 after the capacitive current has decayed. Graphs 21a and 21b show the pulses on line 21 for the alternative integration periods, including part and nearly all of the Off-period respectively. Graphs 54a and 54b show the corresponding integration periods.

The instrument described above was used with commercially available sensors to determine the level of oxygen dissolved in water. Various time intervals for On-period and Off-period were tested and comparative tests were performed with an On-period of 1 second and an Off-period of 1 minute. A galvanic sensor, Rexnord Model 62 (Rexnord Instrument Products, Malvern, Pennsylvania, USA) was used with the instrument and operated with cathode potential zero for the On-period and +0.7 volts for the Off-period. A voltammetric sensor, IBC Model 526-001 (International Biophysics Corporation, Irvine, California, USA) was also used with the instrument, and it was operated at a cathode potential of -0.8 volts for the On-period and



-0.2 volts for the Off-period. It was found that for the Rexnord sensor in air-saturated water, the cathode current, integrated in accordance with the present invention, had a capacitive component of only 5 percent of the integral of the On-period cathode current, yet for non-integrated current measurements at the end of the On-period, the capacitive component was approximately 90 percent of the total cathode current. The corresponding percentages for the IBC sensor were between 1 and 2 percent, and 30 percent. These results show that the instrument of the present invention nearly cancels the contribution of the capacitive component of the cathode current, thus providing, for a given pulse width, much lower "zero" readings than those obtainable with the well known technique of pulse amperometry, thereby improving the accuracy of measurement of oxygen content in water samples.

Another feature of the present invention is the reduction of the magnitude of the capacitive component of the electrode current that is achieved by adding to the electrolyte suitable capillary-active substances such as higher alcohols (for example, capryl alcohol) in order to reduce the amount of capacitive charge to be cancelled. The presence of this additive permits more accurate cancellation of the capacitive charge and, due to quicker transfer of capacitive charge, a narrower pulse width and a shorter integration period.

A particularly beneficial feature of the present invention is that it may be used to measure oxygen level with a cathode material of high peroxide-decomposing ability such as silver or platinum. The present invention reduces measurement errors caused by peroxide accumulation near the cathode.

It will be seen by those skilled in this art that the present invention has provided an instrument and a technique whereby significant improvements in the

measurement of analyte level can be achieved. However, it will also be appreciated by those skilled in the art that many minor variations can be made to the particular embodiment of the instrument that has been described without departing from the essential characteristics of the present invention.

INDUSTRIAL APPLICABILITY

It has been seen that the present invention is particularly useful in the measurement of oxygen levels in water. However, the present invention may also be used for the measurement of the concentration of other analytes, when that measurement involves finding the Faradaic indicator electrode current which flows during a potential pulse in the electrolytic cell and there is a significant capacitive component at the end of the pulse, provided that a substantial proportion of this component is not due to changes in indicator electrode area during the integration period.

CLAIMS

1. An instrument for measuring the concentration of electroactive material, said instrument being adapted for use with an electrolytic cell (14) and comprising means (13) to apply sequentially to the cell an On-potential for an On-period of time and an Off-potential for an Off-period of time, characterised in that said apparatus includes
   a) integration means (18) to integrate the cell current during the application of the On-potential and the Off-potential for a period of time comprising the On-period and at least part of the Off-period;
   b) control means (10) to control the length of the On-period and the Off-period and to preselect the values of the On-potential and the Off-potential; and
   c) measuring means (20) to display and/or record the integrated cell current.

2. An instrument as defined in claim 1, further characterised in that said integration means (18) integrates an applied voltage signal, and a current amplifier (16) is interposed between said cell (14) and said integration means (18), said current amplifier being adapted to amplify said cell current and produce a voltage output signal, proportional to the amplified cell current, to said integration means (18).

3. An instrument as defined in claim 1 or claim 2, further characterised in that said measuring means (20) comprises a readout device, controlled by signals from said control means

(10), said readout device including means to perform analogue to digital conversion and display means to display the said integrated electrode current.

4. An instrument as defined in claim 1, claim 2 or claim 3, in which said integration means (18) is a digital integrator.

5. An instrument as defined in claim 1, claim 2 or claim 3, further characterised in that said voltage application means (13) comprises first and second voltage sources (38,52), connected by respective first and second switches (32,34) to a buffer amplifier (44), said first voltage source (38) being the source of said On-potential and said first switch (32) being connected directly to an input from said control means (10), said second voltage source (52) being the source of said Off-potential and said second switch (34) being connected to said input from said control means (10) via a signal inverter (36), whereby, in the absence of a signal from said control means (10) to apply the On-potential, the Off-potential comprises the output of the buffer amplifier (44).

6. An instrument as defined in claim 1, claim 2 or claim 3, further characterised in that said voltage application means (13) comprises:

a) a source (31) of reference voltage;

b) a potential divider circuit having two arms, one arm consisting of a switch (33) connected to an input to said voltage application means from said control means (10), and first and second resistors

(35,39) in series with said switch (33), the other arm consisting of a third resistor (37) connected to one input of an amplifier (45), the output of said amplifier (45) constituting the output of said voltage application means, said second resistor (39) being in the feedback loop of said amplifier (45) and a connection existing between the said one input of said amplifier (45) and the junction between said first resistor (35) and said second resistor (39);

whereby, when said switch (33) is turned on by an input signal from said control means (10), the output of said amplifier (45) is the On-potential, and at other times the output of said amplifier (45) is the Off-potential.

7. A method of electro-analysis to determine the concentration of electroactive material in an electrolytic cell (14), said method comprising sequentially applying an On-potential to the cell (14) for an On-period of time and applying an Off-potential for an Off-period of time, characterised by the steps of

a) selecting said On-potential to produce a measurable Faradaic cell current and selecting said Off-potential to produce zero Faradaic current in said cell; and

b) integrating the total cell current during the application of the On-potential and the Off-potential for a period of time comprising the On-period and at least part of the Off-period, so that the capacitive

component of the integral of the On-period cell current is cancelled by the integration of the Off-period cell current.

8. A method as defined in claim 7, in which the Faradaic Off-potential current is not zero, said method being characterised in that said integration step comprises

a) a first integration of the cell current as recited in step (b) of claim 7;

b) a second integration of the cell current during the Off-period for a period of time beginning when the capacitive component has decayed to an insignificant value but of time equal to the time of the first integration; and

c) subtracting value of the second integration from the value of the first integration.

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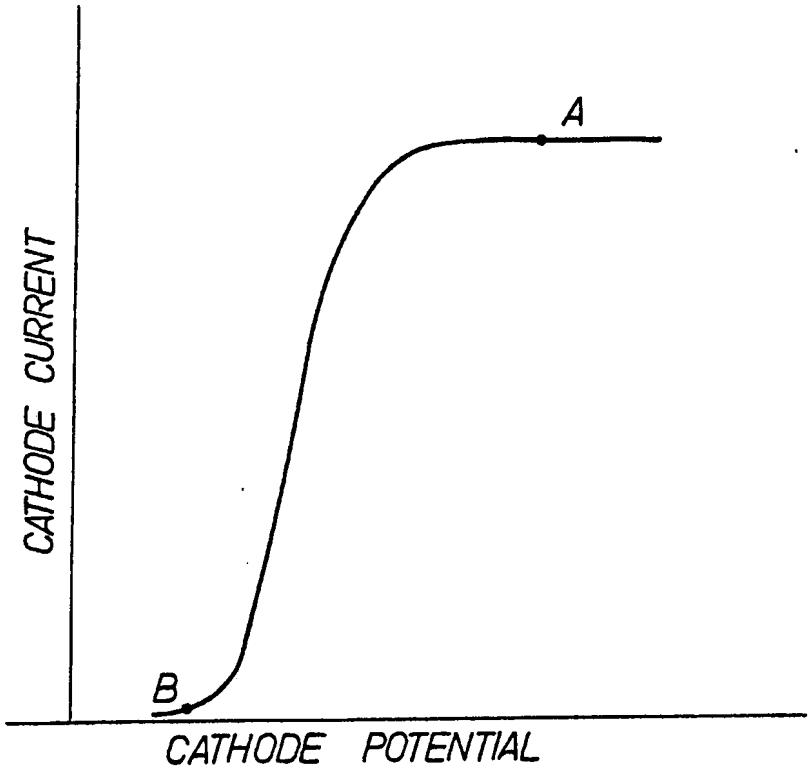

FIG. 1.

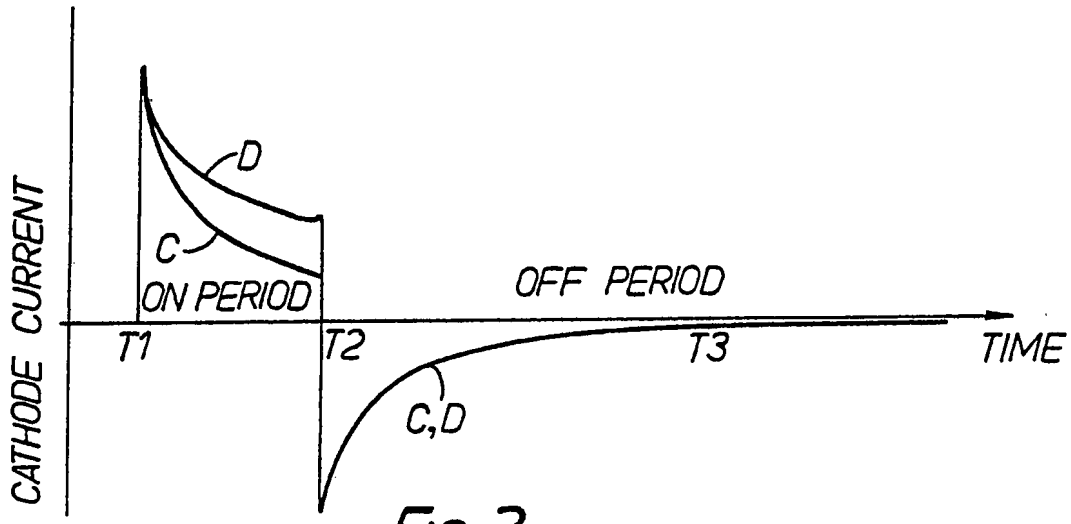

FIG. 2.

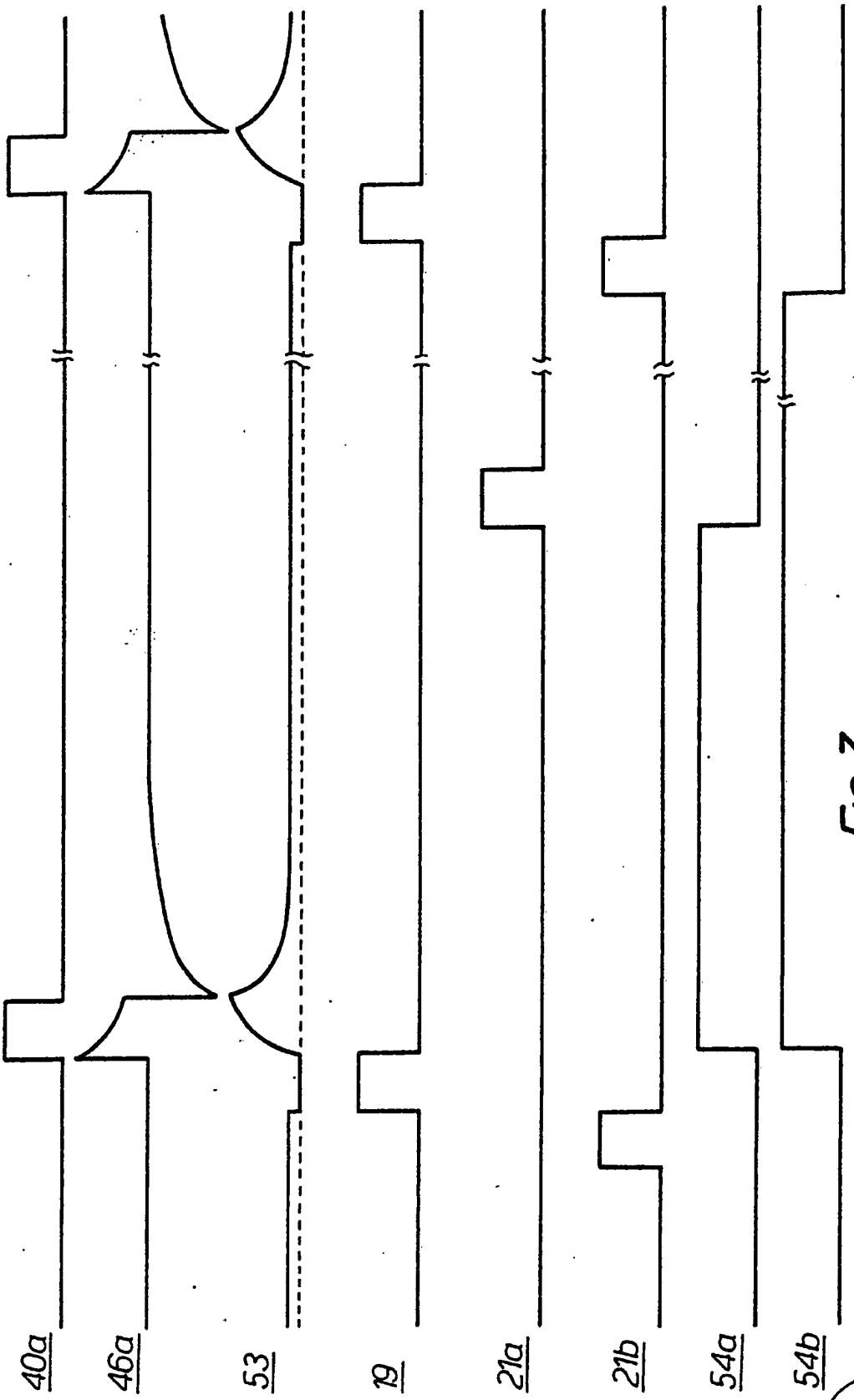

FIG. 3.

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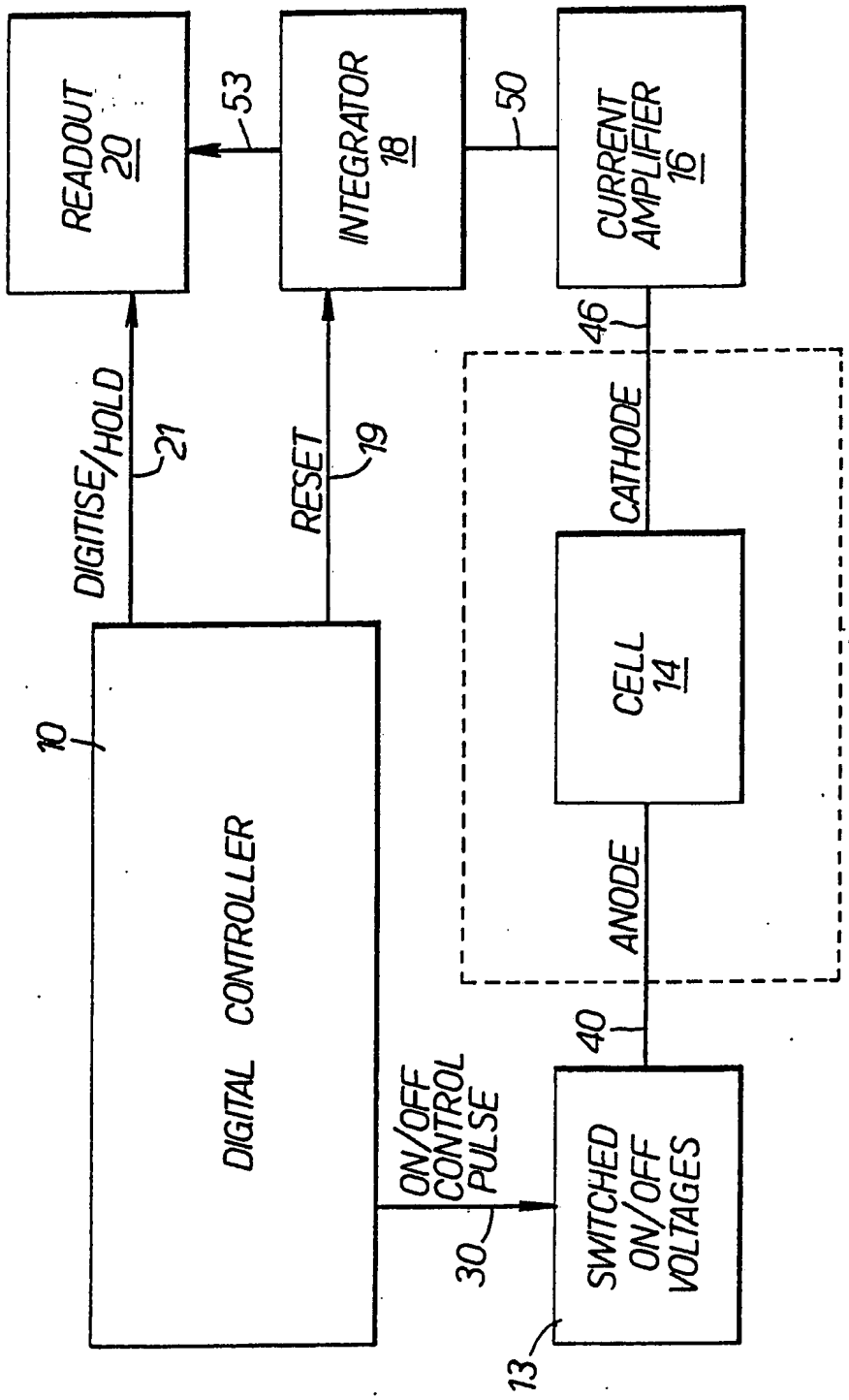

FIG. 4.

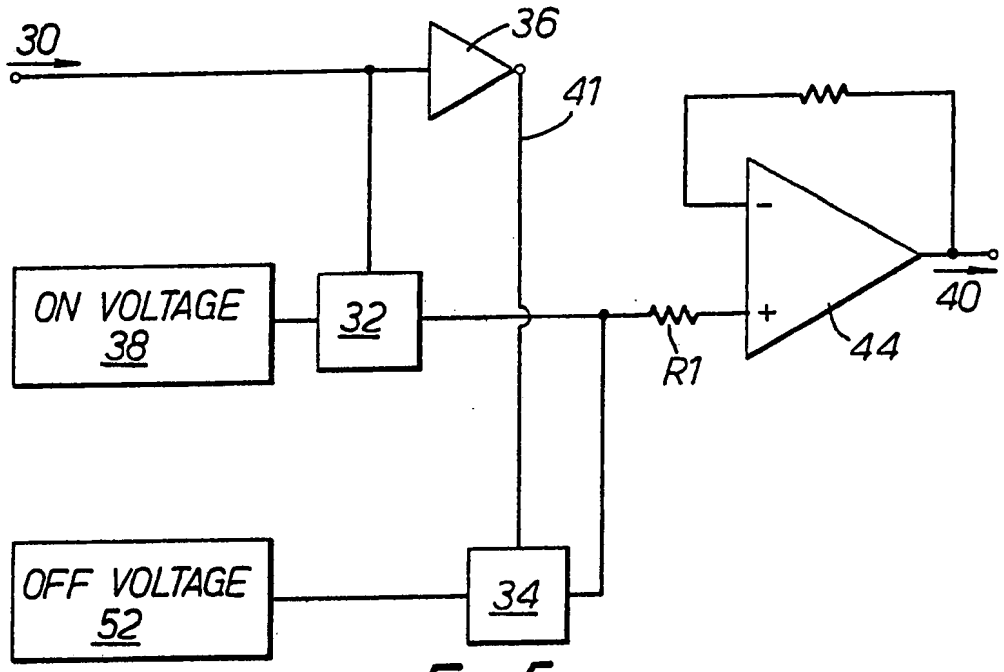

FIG. 5.

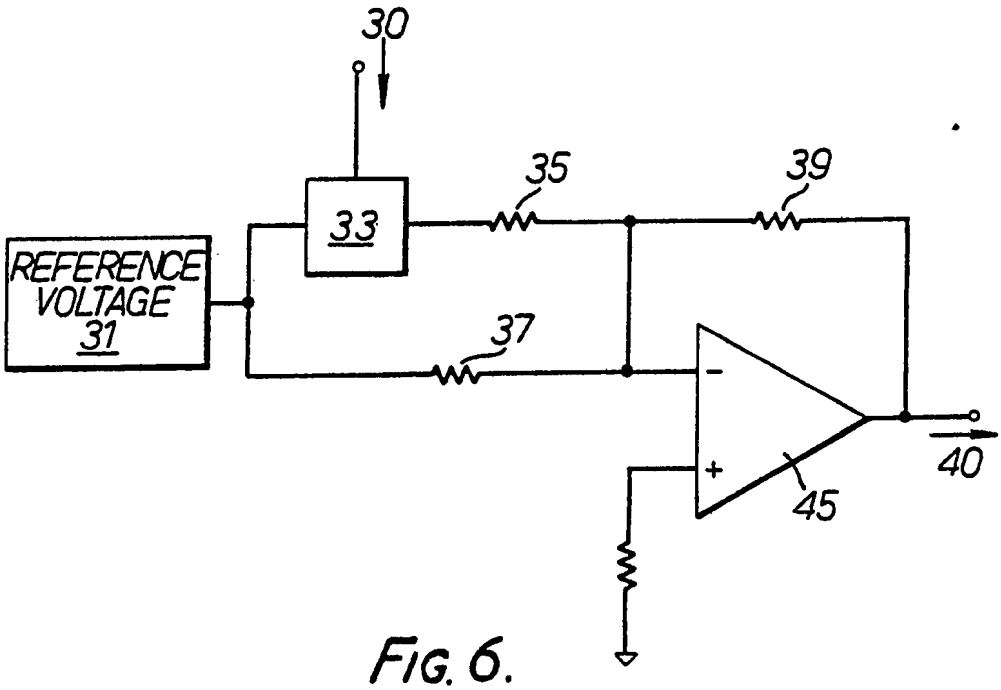

FIG. 6.

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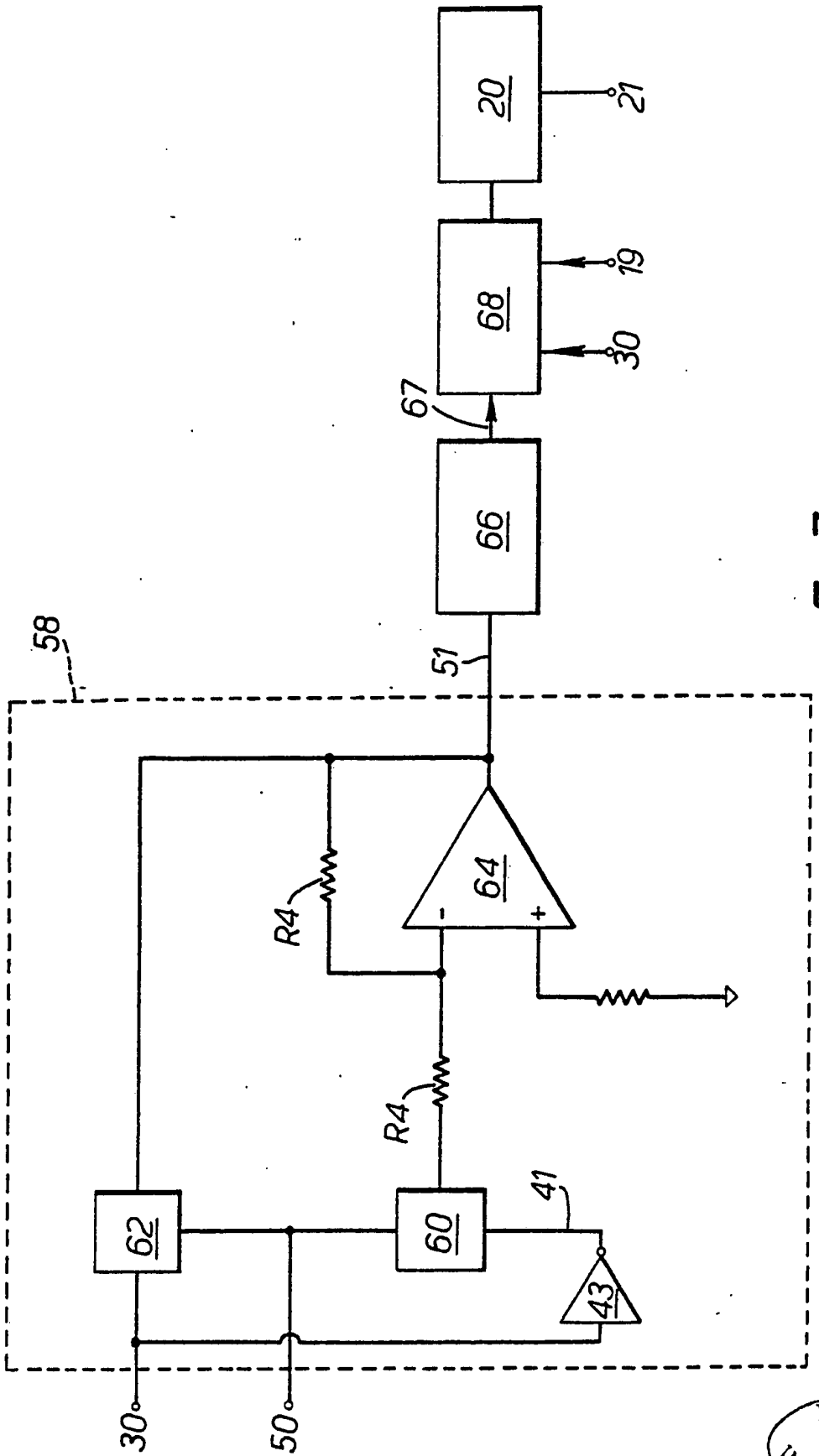

FIG. 7.

INTERNATIONAL SEARCH REPORT

International Application No PCT/AU 82/00175

I. CLASSIFICATION OF SUBJECT MATTER (if several classification symbols apply, indicate all) [3]

According to International Patent Classification (IPC) or to both National Classification and IPC

IPC [3] G01N 27/46

II. FIELDS SEARCHED

Minimum Documentation Searched [4]

Classification System	Classification Symbols
IPC 3	G01N 27/46, 27/48, 27/50, 27/52, 27/54, 27/56, 27/58

Documentation Searched other than Minimum Documentation to the Extent that such Documents are included in the Fields Searched [5]

Aust. Cl. AS ABOVE

III. DOCUMENTS CONSIDERED TO BE RELEVANT [14]

Category *	Citation of Document, [16] with indication, where appropriate, of the relevant passages [17]	Relevant to Claim No. [18]
X	US, A, 2766423 (BARKER, G.C.) 9 OCTOBER 1956 (09.10.56) (& GB, A, 709826)	1-4
A	US, A, 3420764 (SCHLEIN, H.) 7 JANUARY 1969 (07.01.69)	
A	GB, A, 1032231 (SANKYO COMPANY LIMITED) 8 JUNE 1966 (08.06.66)	
A	US, A, 4260680 (MURAMATSU, K. ET AL) 7 APRIL 1981 (07.04.81) (& DE, A, 2845820 & JP, A, 54060996 & FR, A, 2406825 & GB, A, 2009937)	
A	US, A, 4048041 (DAVID, D.J. ET AL) 13 SEPTEMBER 1977 (13.09.77)	
A	US, A, 4105523 (STOLARCZYK, L.G.) 8 AUGUST 1978 (08.08.78)	

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IV. CERTIFICATION

Date of the Actual Completion of the International Search [2]	Date of Mailing of this International Search Report [2]
19 JANUARY 1983 (19.01.83)	25 JANUARY 1983 (25-01-1983)
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